

D. SORPTION DATA RECOMMENDED FOR PERFORMANCE ASSESSMENT

Sorption is a function of water chemistry and the type of tuff at Yucca Mountain. The water chemistry at Yucca Mountain was reviewed by Meijer (1992) and is discussed in “Yucca Mountain Waters” (Section I.C) and “Groundwater Chemistry Model” (Chapter II) of this report. The concentration of the major cations and anions in unsaturated-zone (UZ) groundwaters appears to be intermediate between the saturated-zone tuffaceous waters (for example, from Well J-13) and waters from the Paleozoic carbonate aquifer (from Well UE-25 p#1). Consequently, the first assumption made for the performance-assessment recommendations was that the waters from Wells J-13 and UE-25 p#1 bound the chemistry of the groundwaters at Yucca Mountain.

The second assumption made dealt with grouping all strata on the basis of rock type (the stratigraphy considered from the repository horizon to the accessible environment is outlined in Table 4, page 17). This assumption reduced the number of sorption-coefficient distributions elicited to four per radionuclide: iron oxides, devitrified tuff, vitric tuff, and zeolitic tuff. The basis for this grouping is the fact that sorption of radionuclides is the result of a chemical reaction between the radionuclide in the groundwater and the minerals in the tuff. The mineralogy of the different strata of the same rock group is very similar, and the sorption coefficients can be grouped in terms of these rock types (Thomas 1987).

The containers to be used in the repository were added to the list after consideration of whether the corrosion by-products of the massive multi-purpose container could become a substrate for sorption. Actinides are sorbed strongly by iron oxides. However, although hematite is found in the tuffs at Yucca Mountain, the iron-oxide minerals in the tuffs appear to be “passivated”—that is, all of the sorption sites could be occupied by other metals (Triay et al. 1993b)—and the sorption of the

radionuclides onto tuff (containing iron oxides as trace minerals) is not as large as predicted on the basis of the sorption of radionuclides onto synthetic pure iron oxides. Because the sorption sites on the degraded container material would not necessarily be occupied by other metals, the experts agreed to add iron oxides to the list of “rock” types.

The effect of temperature on sorption coefficients was reviewed by Meijer (1990). Measured sorption coefficients onto tuffs were higher at elevated temperatures for all elements studied: americium, barium, cerium, cesium, europium, plutonium, strontium, and uranium. Consequently, the third assumption made was that sorption coefficients measured at ambient temperatures should be applicable and generally conservative when applied to describing aqueous transport from a hot repository. (This assumption is meaningful provided that the high temperatures that will be sustained for long time periods due to potential high thermal loads do not result in changes in the mineralogy and the water chemistry at Yucca Mountain that are not predictable by short-term laboratory and field experiments.)

Tables 25 shows the parameters for the sorption-coefficient-probability models recommended for performance assessment for the unsaturated-zone units, and Table 26 shows the same parameters for saturated-zone units. We now discuss the source of these values for each of the elements separately.

Americium

Americium sorbs strongly to most materials (Triay et al. 1991b). The potential mechanisms for actinide sorption onto mineral surfaces has been reviewed by Meijer (1992). The sorption-coefficient distributions for americium in Yucca Mountain tuffs and iron oxides given in Tables 25 and 26 were inferred from the data presented by Thomas (1987), Triay et al. (1991b), and Meijer (1992).

Plutonium

One of the problems of interpreting sorption data for plutonium is that this element can exist in multiple oxidation states under oxidizing conditions at near-neutral pH values (Nitsche et al. 1993a). Plutonium can also exist as a polymer (Triay et al. 1991a). The lack of information on the speciation of plutonium in the groundwaters at Yucca Mountain makes it difficult to assess the sorption mechanism for this element. However, the empirical data obtained in Yucca Mountain tuffs indicate that plutonium sorbs strongly. The sorption-coefficient distributions for plutonium in Yucca Mountain tuffs given in Tables 25 and 26 were inferred from the data presented by Thomas (1987) and Meijer (1992).

Uranium

No additional data for uranium has been collected for Yucca Mountain tuffs since the 1991 total-system performance-assessment effort (TSPA-1991). Consequently, no change was made for the sorption-coefficient distributions used for this element. As previously discussed (Meijer 1992), uranium sorbs strongly to synthetic iron oxides.

Thorium

The information elicited for americium was also used for thorium. This approach is due both to the lack of sorption information available for thorium and to the similarities exhibited by the sorption behavior of these two elements (Thomas 1987).

Radium

Barium has been used as an analog for radium in the experiments performed at Los Alamos (Thomas, 1987). These elements sorb to Yucca Mountain tuffs via an ion-exchange mechanism and surface-adsorption reactions (Meijer 1992). The sorption-coefficient distributions for radium in Yucca Mountain tuffs and iron oxides given in Tables 25 and 26 were inferred from the data pre-

sented by Thomas (1987), Meijer (1992), and Triay et al. (1991c).

Lead

Lead tends to complex with fulvics in the groundwaters and sorbs as a complex. The sorption-coefficient distributions for lead in Yucca Mountain tuffs and iron oxides given in Tables 25 and 26 were inferred from the data presented by Meijer (1990).

Neptunium

Sorption-coefficient distributions for neptunium in tuff are the same as those used in TSPA-1991. Recently obtained data (Triay et al. 1993b) agrees with previous observations. Neptunium is a poorly sorbing radionuclide in tuff even when the tuffs are known to have iron oxides, because the iron oxides in the tuff appear to be passivated. The neptunium sorption-coefficient distribution for sorption onto iron oxides given in Tables 25 and 26 was inferred from data presented by Meijer (1992) and Triay et al. (1993b) for sorption onto synthetic iron oxides.

Protactinium

Very little information exists for protactinium sorption onto tuffs (Thomas 1987), so the experts decided to use for this element the same sorption coefficients elicited for neptunium.

Tin

There is very little information for the sorption of tin onto tuffs (Thomas 1987). Based on the data available, Meijer (1992) suggested that tin exhibited large values of K_d in the devitrified tuffs (larger than 1000 ml/g). The sorption-coefficient distributions given in Tables 25 and 26 were inferred from the work by Andersson (1988); the uniform distributions chosen was the result of the expert's uncertainty about the sorption of tin.

Table 25. Sorption-coefficient Distributions for Unsaturated-zone Units

Element	Rock type	Min K_d (ml/g)	Max K_d (ml/g)	$E[x]$	COV*	Distribution type
Americium	Devitrified	100	2000	400	0.20	Uniform
	Vitric	100	1000			Beta
	Zeolitic	100	1000			Uniform
	Iron oxide	1000	5000			Uniform
Plutonium	Devitrified	20	200	100	0.25	Beta
	Vitric	50	200	100	0.25	Beta
	Zeolitic	30	200	100	0.25	Beta
	Iron oxide	1000	5000			Uniform
Uranium	Devitrified	0	4.0	2.0	0.3	Beta
	Vitric	0	3.0	1.0	0.3	Beta
	Zeolitic	0	30.0	7.0	1.0	Beta(exp)
	Iron oxide	100	1000			Uniform
Neptunium	Devitrified	0	6.0	1.0	0.3	Beta
	Vitric	0	15.0	1.0	1.0	Beta(exp)
	Zeolitic	0	3.0	0.5	0.25	Beta
	Iron oxide	500	1000			Uniform
Radium	Devitrified	100	500	30	1.0	Uniform
	Vitric	50	100			Uniform
	Zeolitic	1000	5000			Uniform
	Iron oxide	0	500			Beta(exp)
Cesium	Devitrified	20	1000	30	1.0	Uniform
	Vitric	10	100			Uniform
	Zeolitic	500	5000			Uniform
	Iron oxide	0	500			Beta(exp)
Strontium	Devitrified	10	50	10	0.25	Uniform
	Vitric	0	20			Uniform
	Zeolitic	500	2000			Uniform
	Iron oxide	0	30			Beta
Nickel	Devitrified	0	500	100	0.33	Beta
	Vitric	0	100	50	0.33	Beta
	Zeolitic	0	500	100	0.33	Beta
	Iron oxide	0	1000			Uniform
Lead	Devitrified	100	500			Uniform
	Vitric	100	500			Uniform
	Zeolitic	100	500			Uniform
	Iron oxide	100	1000			Uniform
Tin	Devitrified	20	200			Uniform
	Vitric	20	200			Uniform
	Zeolitic	100	300			Uniform
	Iron oxide	0	5000			Uniform
Protactinium	Devitrified	0	100			Uniform
	Vitric	0	100			Uniform
	Zeolitic	0	100			Uniform
	Iron oxide	500	1000			Uniform
Selenium	Devitrified	0	30	3	1.0	Beta(exp)
	Vitric	0	20	3	1.0	Beta(exp)
	Zeolitic	0	15	2	1.0	Beta(exp)
	Iron oxide	0	500	30	1.0	Beta(exp)
Carbon	Iron oxide	10	100			Uniform
Actinium, Niobium, Samarium, Thorium, Zirconium: see Americium						
Chlorine, Technetium, Iodine		0	0			

*Coefficient of variation: $COV = \sigma[x]/E[x]$

Table 26. Sorption-coefficient Distributions for Saturated-zone Units

Element	Rock type	Min K_d (ml/g)	Max K_d (ml/g)	$E[x]$	COV*	Distribution type
Americium	Devitrified	100	2000	400	0.20	Uniform
	Vitric	100	1000			Beta
	Zeolitic	100	1000			Uniform
	Iron oxide	1000	5000			Uniform
Plutonium	Devitrified	50	300	100	0.15	Beta
	Vitric	50	300	100	0.15	Beta
	Zeolitic	30	300	100	0.15	Beta
	Iron oxide	1000	5000			Uniform
Uranium	Devitrified	0	5.0	2.0	0.3	Uniform
	Vitric	0	4.0	1.0	0.3	Uniform
	Zeolitic	5	20.0	7.0	0.3	Beta
	Iron oxide	100	1000			Uniform
Neptunium	Devitrified	0	10.0	3.0	0.3	Beta
	Vitric	0	15.0	1.5	1.0	Beta(exp)
	Zeolitic	0	12.0	4.0	0.25	Beta
	Iron oxide	500	1000			Uniform
Radium	Devitrified	100	500	30	1.0	Uniform
	Vitric	100	500			Uniform
	Zeolitic	1000	5000			Uniform
	Iron oxide	0	1500			Beta(exp)
Cesium	Devitrified	20	1000	30	1.0	Uniform
	Vitric	10	100			Uniform
	Zeolitic	500	5000			Uniform
	Iron oxide	0	500			Beta(exp)
Strontium	Devitrified	10	200	10	0.25	Uniform
	Vitric	20	50			Uniform
	Zeolitic	2000	5000			Log uniform
	Iron oxide	0	30			Beta
Nickel	Devitrified	0	500	100	0.33	Beta
	Vitric	0	200	100	0.33	Beta
	Zeolitic	0	500	100	0.33	Beta
	Iron oxide	0	1000			Uniform
Lead	Devitrified	100	500			Uniform
	Vitric	100	500			Uniform
	Zeolitic	100	500			Uniform
	Iron oxide	100	1000			Uniform
Tin	Devitrified	20	200			Uniform
	Vitric	20	200			Uniform
	Zeolitic	100	300			Uniform
	Iron oxide	0	5000			Uniform
Protactinium	Devitrified	0	100			Uniform
	Vitric	0	100			Uniform
	Zeolitic	0	100			Uniform
	Iron oxide	500	1000			Uniform
Selenium	Devitrified	0	30	3	1.0	Beta(exp)
	Vitric	0	20	3	1.0	Beta(exp)
	Zeolitic	0	15	2	1.0	Beta(exp)
	Iron oxide	0	500	30	1.0	Beta(exp)
Carbon	Iron oxide	10	100			Uniform
Actinium, Niobium, Samarium, Thorium, Zirconium: see Americium						
Chlorine, Technetium, Iodine		0	0			

*Coefficient of variation: $COV = \sigma[x]/E[x]$

Nickel

For devitrified, vitric, and zeolitic tuffs, the nickel sorption-coefficient distributions given in Tables 25 and 26 were inferred from data presented by Meijer (1992). For iron oxides, the nickel sorption-coefficient distribution was inferred from the data presented by Siegel et al. (1992 and 1993).

Cesium

Cesium sorption-coefficient distributions for tuff and iron oxides were inferred from the data presented by Thomas (1987), Meijer (1992), and Triay et al. (1991c). Cesium has one of the highest selectivity coefficients for zeolites among all chemical elements (Meijer 1992). Cesium sorption onto devitrified and vitric samples could be the result of ion exchange onto clays or feldspars in the tuff samples or surface-adsorption reactions (Meijer 1992).

Strontium

Strontium sorption-coefficient distributions for tuff and iron oxides were inferred from the data presented by Thomas (1987) and Triay et al. (1991c). Strontium sorbs strongly onto zeolites by ion exchange. This element's sorption onto other types of tuff may be dominated by the amount of clay in the tuff units. The values given in Tables 25 and 26 are generally conservative.

Selenium

There are limited data on tuff for selenium sorption (Thomas 1987), so the experts decided to use the same sorption-coefficient distributions for selenium as the ones elicited for uranium. This decision is a conservative one because uranium can be oxidized much more readily than selenium in Yucca Mountain groundwaters.

Carbon

Carbon is a special case because transport is

expected to occur primarily in the gaseous phase as carbon dioxide. The major retardation mechanism is exchange of carbon-14 with the carbon in the carbon dioxide dissolved in the groundwater.

Actinium, Samarium, Niobium, and Zirconium

All these elements are strongly sorbing (Meijer 1992). The experts advised using for these elements the same sorption-coefficient distributions as those elicited for americium.

Iodine, Technetium, and Chlorine

Iodine and chlorine have anions that do not sorb onto tuffs. Technetium exists as pertechnetate under oxidizing conditions and does not sorb either (Triay et al. 1993a).